Polarized neutron diffraction studies of Gd-Y synthetic superlattices

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The microscopic magnetic structures of coherent, single-crystal epitaxially grown superlattices consisting of successive bilayers of $N_{\rm Gd}$ basal planes of hexagonal-close-packed Gd followed by $N_{\rm Y}$ such atomic planes of nonmagnetic Y have been studied by polarized neutron diffraction. It has been found that for $N_{\rm Gd} = 10$, either a simple parallel alignment of the ferromagnetic Gd layers or an antiphase domain structure occurs, depending on the number of intervening Y planes in an oscillatory manner. These data are consistent with a RKKY coupling mechanism between Gd layers. In addition to the investigation of the interlayer interactions, measurements of the magnetization profile across the thickness of a Gd layer have been performed.

INTRODUCTION

Now that single-crystal rare-earth (RE) superlattices can be grown with a high degree of perfection and limited interdiffusion,[1] neutron diffraction studies of fundamental magnetic phenomena including interlayer interactions and the effects of reduced dimensionality can be performed in a controlled and systematic manner. The results of a neutron diffraction investigation of $(\mathrm{Gd}_{N_{\rm Gd}}\text{-}\mathrm{Y}_{N_{\rm Y}})_M$ superlattices composed of M successive bilayers of $N_{\rm Gd}$ basal planes of hexagonal-close-packed Gd followed by $N_{\rm Y}$ such planes of nonmagnetic Y will be described here. Details of the preparation of the Gd-Y superlattices are reported elsewhere.[1,2]

The neutron diffraction measurements were performed at the high-flux beam reactor at Brookhaven National Laboratory and the high-flux isotope reactor at Oak Ridge National Laboratory. Triple-axis spectrometers in the elastic scattering mode with polarizing monochromator and analyzer (either $\mathrm{Cu_2MnAl}$ or $^{57}\mathrm{Fe}$ crystals) and neutron spin flippers before and after the sample were used so that the four neutron spin-dependent scattered intensities, $I^{++}(Q)$, $I^{--}(Q)$, $I^{+-}(Q)$, and $I^{-+}(Q)$ could be measured[3] (the two superscripts denote the initial and final neutron spin states and $|\mathbf{Q}| = |\mathbf{k}_f - \mathbf{k}_i|$, where $\mathbf{k}_f$ and $\mathbf{k}_i$ are the final and initial neutron wave vectors, respectively). Satellite reflections due to the chemical and magnetic modulations of the superlattice were observed and found to have intrinsic widths corresponding to a spatial coherence of the order of ten bilayer periods along c^* for all of the samples examined.

INTERLAYER INTERACTIONS

In the case of the metallic, magnetic rare earths, the magnetic moments are well localized and the indirect exchange interaction is via the conduction electrons. The long-range nature of this Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction can be expected to give rise to a modulation of the magnetic properties in an artificially layered structure. Three Gd-Y superlattice samples deposited on approximately 1-in. square substrates, each with the same number of Gd planes per bilayer, were studied by neutron diffraction[4]: $(\mathrm{Gd}_{10}\text{-}\mathrm{Y}_6)_{189}$, $(\mathrm{GD}_{10}\text{-}\mathrm{Y}_{10})_{225}$, and $(\mathrm{Gd}_{10}\text{-}\mathrm{Y}_{20})_{100}$.

The individual Gd layers of the superlattices become ferromagnetic at about 285 K with the moments lying in the basal planes (for fields of the order of several hundred Oe or more applied perpendicular to c^*). Above T_c, satellite reflections are observed for all three samples about the $(00l)$ Bragg peaks at positions along the $[00l]$ direction corresponding to integer multiples of $2\pi/\lambda_{\rm SL}$, where $\lambda_{\rm SL}$ is the bilayer thickness (up to five higher-order harmonics are easily observable). These reflections arise solely from the nuclear scattering associated with the chemical modulation so that $I^{++} = I^{--}$. Below T_c, with the sample in a field of the order of a hundred Oe applied perpendicular to $\mathbf{Q}(\mathbf{Q}\|c^*)$, it is found that $I^{++} \neq I^{--}$ which indicates the presence of an additional ferromagnetic component.

For the superlattices with $N_{\rm Y} = 6$ and 20, all of the Gd layers are aligned with one another in a simple ferromagnetic array. The behavior of the superlattice with $N_{\rm Gd} = N_{\rm Y} = 10$, however, is quite different. In addition to the magnetic non-spin-flip (NSF) scattering which occurs at the satellites positioned at multiples of $2\pi/\lambda_{\rm SL}$ about $(00l)$ below T_c, another set of satellites appears at odd integer multiples of $2\pi/2\lambda_{\rm SL}$ corresponding to a *doubling* of the chemical bilayer periodicity. The scattering associated with these additional satellite reflections is entirely spin-flip (SF) in nature. Furthermore, a field of several thousand Oe

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applied in the plane of the substrate is sufficient to reduce the intensities of the SF satellites to zero. The measured volume-integrated intensities were compared to various models for the magnetic structure, incorporating the chemical composition (interdiffusion) and atomic-plane-spacing (strain) modulations along the c axis which were deduced from x-ray measurements.[5] It is found that an antiphase domain structure or simple antiferromagnetic arrangement of the Gd *layers* (again, each layer consisting of $N_{\rm Gd}$ ferromagnetically aligned atomic planes) gives the best agreement with the data.[4]

One particularly appealing explanation to account for the observed oscillatory behavior is to attribute the coupling between two Gd layers across an intervening Y layer to the RKKY interaction. In order to estimate the strength and sign of this coupling, a calculation of the interaction between two Gd monolayers separated by pure Y (for which the calculated susceptibility function was assumed[6]) was performed.[7] While the validity of this approximation has not been rigorously tested, it is probably the simplest physically plausible computation that can be made short of doing a superlattice band calculation. The sign and relative strength of the interaction obtained from this calculation are found to be consistent with the data discussed above and magnetization data for five additional superlattice samples which show that the remanence and saturation field have the same oscillatory dependence on $N_{\rm Y}$.[8]

MAGNETIZATION PROFILE

Another interesting application of polarized neutron diffraction to the study of magnetic superlattices is the determination of the magnetization profile across the thickness of a thin ferromagnetic layer. Theoretical studies[9] of the magnetic critical behavior at a vacuum-surface interface have even predicted, for example, that for sufficiently enhanced coupling in the surface layer there can be distinct transitions in the surface and in the underlying bulk. Recently, surface-enhanced magnetic order and magnetic surface reconstruction on Gd (001) has been reported.[10] In the case of a single-crystal superlattice it is in fact possible, in principle, to determine the magnetization of each of the N atomic planes within a layer by neutron diffraction. If the magnetization profile is then measured as a function of temperature, the critical exponents for the magnetization of each atomic plane, including that at the interface, can be deduced and the effects of reduced dimensionality and interface anisotropy inferred.

For a ferromagnet which is saturated in a magnetic field applied perpendicular to $\mathbf{Q}$, the difference ΔI_m in the non-spin-flip scattered intensities associated with the mth satellite along the $[00l]$ direction is given by

$$\begin{aligned}\Delta I_m &= I_m^{++} - I_m^{--}\\ &= 4C_m(\mathrm{Re}\,F_{N_m}\,\mathrm{Re}\,F_{M_m} + \mathrm{Im}\,F_{N_m}\,\mathrm{Im}\,F_{M_m}),\end{aligned} \tag{1}$$

where Re and Im denote the real and imaginary parts of the nuclear and magnetic structure factors F_N and F_M, respectively, which can be written as

$$F_N = \sum_j (\mathrm{Re}\,b_j - i\,\mathrm{Im}\,b_j)e^{iQu_j} \tag{2}$$

and

$$F_M = \sum_j p_j e^{iQu_j}. \tag{3}$$

In the above equations C_m is a normalization constant, u_j is the position, and b_j and p_j the nuclear and magnetic coherent scattering lengths, respectively, of the jth atomic plane in the unit cell.

The magnetic scattering length p_j is proportional to the magnetization of the jth atomic plane. Equations (1)–(3) can be combined to give

$$\begin{aligned}\Delta I_1 &= \alpha_{11}p_1 + \alpha_{12}p_2 + \cdots + \alpha_{1l}p_l\,,\\ \Delta I_2 &= \alpha_{21}p_1 + \alpha_{22}p_2 + \cdots + \alpha_{2l}p_l\,,\\ &\ \ \vdots\\ \Delta I_m &= \alpha_{m1}p_1 + \alpha_{m2}p_2 + \cdots + \alpha_{ml}p_l\,,\end{aligned} \tag{4}$$

where the coefficients α_{m_l} can be calculated provided that the nuclear scattering lengths, compositional modulation (i.e., interdiffusion), and the atomic plane positions (or strain modulation) can be obtained with sufficient accuracy from x ray and/or neutron diffraction measurements (in the latter case with an applied field strong enough to align the moments parallel to $\mathbf{Q}$). If there are N_M magnetic atomic planes within a bilayer, then N_M satellites must be observed in order to determine the N_M individual atomic plane magnetizations. Of course, if an insufficient number of satellites are observed to obtain each atomic plane magnetization, a model for the magnetization profile represented by a trapezoidal or error function, for example, can still be fit to the data.

In order to study the magnetization profile as a function of temperature, the $(\mathrm{Gd}_{10}\text{-}\mathrm{Y}_{20})_{100}$ sample was selected to minimize the effects of interlayer coupling. The volume-inte-

TABLE I. Comparison of observed diffracted intensities $\Delta I \equiv I^{++}(Q) - I^{--}(Q)$ and corresponding values predicted for the model of the magnetization profile described in the text. The magnetization is nearly uniform over the eight interior Gd planes but decreases more in the interface planes than can be attributed to a simple dilution by alloying alone.

Satellite index	Neutron wavelength (Å)	Q (Å^{-1})	ΔI Observed	ΔI Calculated
−2	2.353	2.040	−185.	−73.
−1	2.353	2.113	1585.	1145.
0	2.353	2.186	41478.	35090.
1	2.353	2.259	−5648.	−4931.
2	2.353	2.331	−550.	−611.
−2	1.651	2.040	964.	1068.
−1	1.651	2.113	4613.	4209.
0	1.651	2.186	32982.	30040.
1	1.651	2.259	−1873.	−1369.
2	1.651	2.331	−8	−72.
−2	1.412	2.040	409.	450.
−1	1.412	2.113	1671.	1622.
0	1.412	2.186	8468.	8734.
1	1.412	2.259	−77.	−47.
2	1.412	2.231	57.	35.
	$T = 15$ K	$H \simeq 100$ Oe	$(\mathrm{Gd}_{10}\text{-}\mathrm{Y}_{20})_{100}$	

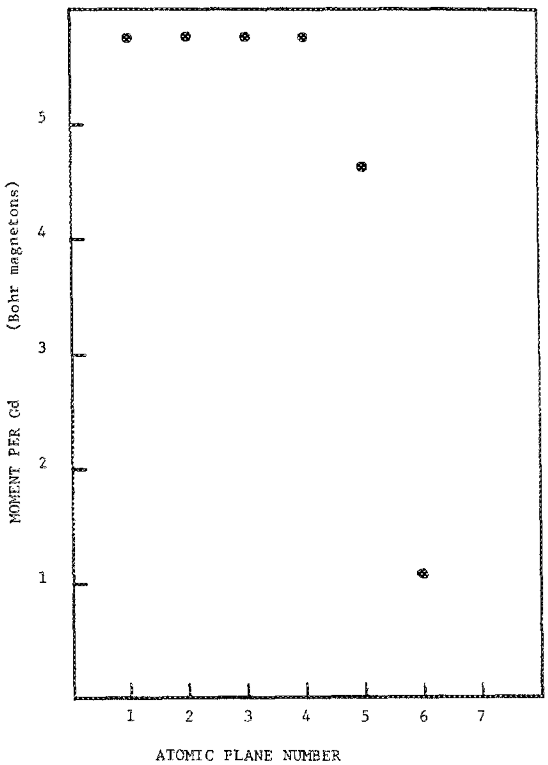

FIG. 1. Magnetization profile of the $(Gd_{10}\text{-}Y_{20})_{100}$ superlattice at 15 K in a field of ≈ 100 Oe as determined by the fit to the neutron data described in the next. The moment per Gd atom is nearly constant in the interior of the Gd layer (atomic plane number 1 is at the center of the layer whereas the center of the interfacial region is between planes 5 and 6). However, the magnetization of the atomic planes in the interfacial region decreases more than can be attributed to a simple dilution by alloying alone (the percentages of Gd atoms in a basal plane were taken to be 100, 100, 95, 80, 50, 50, 20, 5, and 0 for planes 1–9, respectively).

grated intensities of four satellites and the (002) reflection (where $\Delta I_{m=0} = \Delta I_{(002)}$ is proportional to the average magnetization of the planes in a layer) were measured as a function of temperature through T_c at three different neutron wavelengths. For the naturally occuring isotopic mixture of Gd, the real and imaginary parts of the coherent nuclear scattering amplitude change significantly with wavelength over the slow neutron energy range. The wavelength-dependent values of the scattering length were determined from transmission measurements using the Breit–Wigner formulas for resonance scattering to be $0.47 - i1.25$, $0.82 - i1.35$, and $1.1 - i1.30$ (units of 10^{-12} cm) at 2.35, 1.65, and 1.41 Å, respectively. Although the absorption cross section of the natural Gd is relatively high, the actual attenuation of the neutron beam was not prohibitive at high angles because the sample is extremely thin. The magnetic form factor for elemental Gd[11] was assumed and the relatively small changes in absorption and Debye–Waller factors over the limited Q-range were neglected. The normalization constants C_m (defined above) were determined above T_c. Somewhat better fits to the data were obtained if the normalization constants and/or nuclear coherent scattering lengths were treated as adjustable parameters and allowed to vary by several percent. Table I compares the observed intensities with those calculated for a magnetization profile that is nearly uniform with a value of 5.7 μ_B per Gd atom over the eight interior Gd planes, but in which the magnetization of the interface planes decreases more than can be attributed to a simple dilution by alloying alone as shown in Fig. 1. (This model is also consistent with magnetic x-ray diffraction results.[5]) The compositional and strain modulation profiles obtained by x-ray diffraction for a $(Gd_{21}\text{-}Y_{21})$ superlattice were initially assumed.[5] However, it was found that a relatively small adjustment of the amplitude of the strain profile (of the order of 10%) was necessary to obtain a good fit and that the values for ΔI_m (other than $\Delta I_{(002)}$) are in fact quite sensitive to the strain modulation (which is itself strongly temperature dependent). The validity of using the strain modulation profile obtained for a $(Gd_{21}\text{-}Y_{21})$ superlattice may therefore be questionable and further analysis is required before any conclusions regarding the temperature dependence of the magnetization profile can be drawn. A more comprehensive report detailing the analysis and fitting of the neutron data will be forthcoming.

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